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RemarksA. Basis for claim amendments

Basis for the amendment to claim 1 to --20%-- includes page 3, line 22 of the verified translation of the application filed on July 12, 2006.

Basis for the amendment to claim 1 to --acrylic acid-- includes claim 1 itself.

B. The Office Action

Claims 1 and 3-6 are pending.

On page 2 of the Office Action, independent claim 1 and dependent claims 4 and 6 were rejected under 35 U.S.C. 103(a) as being unpatentable over Tenten et al. (US 5,677,261), in view of Neher et al. (US 5,387,720).

On page 4 of the Office Action, dependent claim 3 was rejected under 35 U.S.C. 103(a) as being unpatentable over Tenten et al. (US 5,677,261), in view of Neher et al. (US 5,387,720), and further in view of Unverricht et al. (US 6,403,829).

On pages 4-5 of the Office Action, dependent claim 5 was rejected under 35 U.S.C. 103(a) as being unpatentable over Tenten et al. (US 5,677,261), in view of Neher et al. (US 5,387,720), and further in view of Uchida et al. (US 4,871,700).

C. Applicant's discussionC.1.a. KSR International as to scope and content

It is respectfully submitted that the appropriate standard of review is found in KSR International Co. v. Teleflex Inc. et al., 550 U.S. ____, 127 S. Ct. 1727 (2007), and that this decision mandates that the scope and content of the prior art are to be determined.

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C.1.b. A review of the scope and content of the Tenten et al. and Neher et al. reference in relation to the claimed first and second steps, the claimed starting material, and the claimed final product shows that the primary reference is applied against a second step and the secondary reference is applied against a first step

Claim 1 positively claims a first step. This first step also claims a starting material of an aqueous glycerol solution.

Claim 1 also positively claims a second step. This second step also claims the final product of acrylic acid (not methacrylic acid).

The primary reference, Tenten et al., is being applied against the second step. The Office Action points out that "Tenten et al. teaches the preparation of acrylic acid from acrolein by gas-phase catalytic oxidation (column 11, lines 27-28). As to such a teaching, it is noted that acrolein (not an alcohol and not glycerol) is the starting material.

The secondary reference, Neher et al., is being applied against the first step. For example, the scope and content of the Neher et al. reference includes the disclosure in Example 1 that a 20 wt. % aqueous glycerol solution (water content: 80 wt. %) is pumped at 40 ml/h into a vaporizer to obtain acrolein.

Thus, according to the Office Action, the Neher et al. reference discloses that a glycerol solution may be used to produce acrolein, and the Tenten et al. reference discloses acrolein may be used to produce acrylic acid.

Thus, Tenten et al. discloses not the starting material of glycerol, and not the claimed first step, but only the final product. The secondary reference, Neher et al., does nothing to "modify" or "cure" Tenten et al. The only role

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of the secondary reference is to be cited against the starting material of glycerol and the first step.

All inventions are combinations of old elements. If the present invention is a combination of step A and step B, then the Office Action is citing one reference that may or may not relate to the step B and another reference that may or may not relate to the step A. It is respectfully submitted that KSR International demands more.

Even the Office Action appears to acknowledge that there is no tie between the references of Tenten et al. and Neher et al. For example, on page 5 of the Office Action, it is stated that "the Examiner recognizes that obviousness can only [emphasis added] be established by combining or modifying the teachings of the prior art to produce the claimed invention where there is some teaching, suggestion, or motivation to do so found either in the references themselves or in the knowledge generally available to one of ordinary skill in the art." The Office Action then provides that "In this case, it is permissible for the Examiner to rely on disclosures, which fairly teach embodiments of Applicant's invention. The claims require a multitude of elements and it is reasonable [emphasis added] for one of ordinary skill in the art to consider these elements being used together." Why such is reasonable is not addressed and it is respectfully submitted that KSR International requires at least such.

As asserted in the Amendment and Remarks of August 18, 2007, there is no disclosure in any of the cited references to teach or suggest the continuous integrated process of independent claim 1. Moreover, as discussed below in the next section, at least the Tenten et al. reference teaches away from what is claimed.

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C.2.a. KSR International as to teaching away

On page 12 of the slip opinion, the Supreme Court of the United States in KSR International Co. reaffirms the concept of teaching away:

In *United States v. Adams*, 383 U. S. 39, 40 (1966), a companion case to *Graham*, the Court considered the obviousness of a "wet battery" that varied from prior designs in two ways: It contained water, rather than the acids conventionally employed in storage batteries; and its electrodes were magnesium and cuprous chloride, rather than zinc and silver chloride. The Court recognized that when a patent claims a structure already known in the prior art that is altered by the mere substitution of one element for another known in the field, the combination must do more than yield a predictable result. 383 U. S., at 50-51. It nevertheless rejected the Government's claim that Adams's battery was obvious. The Court relied upon the corollary principle that when the prior art teaches away from combining certain known elements, discovery of a successful means of combining them is more likely to be nonobvious. *Id.*, at 51-52. When Adams designed his battery, the prior art warned that risks were involved in using the types of electrodes he employed. The fact that the elements worked together in an unexpected and fruitful manner supported the conclusion that Adams's design was not obvious to those skilled in the art. [emphasis added]

C.2.b. The Tenten et al. reference teaches away from what is claimed

As to the Tenten reference, the Examiner contends on page 3, third to first lines from bottom of the Final Office Action that "Note that the primary reference, Tenten et al., appears to recognize the equivalency of alkenes and alcohols for gas-phase oxidation (column 12, lines 8-10). The expected result would be the production of acrylic acid from glycerol in high yield."

Applicant respectfully begs to differ with the Examiner. First, applicant discusses how the disclosure pointed out by the Examiner (column 12, lines 8-10) is different from what is claimed. Second, applicant shows how such a disclosure and the Tenten et al. reference as a whole teaches away from what is claimed.

(1) In the Tenten et al. reference, namely in column 12, lines 8-10, the alcohol which is used as a starting

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material is tert-butanol, and the final product (where alcohol is used as a starting material) is methacrylic acid. (It is noted that the Tenten et al. reference teaches acrylic acid as a final product in column 11, line 27, but the starting material in this case is acrolein.) On the other hand, in the present invention, the alcohol which is used as a starting material is glycerol, and the final product is acrylic acid. Therefore, both the claimed starting material and the claimed final product in the present invention differ from those in the Tenten et al. reference.

(2) JP-A-59-210031 (a copy and a partial English translation are enclosed herewith) discloses that an alcohol makes a dehydration reaction to form an olefin (alkene) (please refer to claim 1 of the above partial English translation of the JP reference). Particularly, the JP reference discloses in Examples 1 and 2 that tert-butanol makes a dehydration reaction to form isobutene (please refer to Examples 1 and 2 of the above partial English translation of the JP reference). Therefore, it appears that, also in the Tenten et al. reference, tert-butanol which is used as the alcohol being a starting material makes a dehydration reaction to form isobutene in the same way, and thereafter this isobutene makes an oxidation reaction to form methacrylic acid. On the other hand, as to the present invention, if an olefin, namely, propene, according to the above teaching of the JP reference were formed in the step of the dehydration reaction of an alcohol, namely, glycerol and if this propene according to the teaching of column 4, lines 7-30 of the Tenten et al. reference made an oxidation reaction to form acrylic acid via acrolein, then a catalyst for the oxidation reaction of propene would be necessary separately. However, in the present invention, such a

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catalyst is not necessary. Therefore, the Tenten et al. reactions not only differ from what is claimed, but actually teach away from what is claimed.

C.3. In view of concerns of the Examiner, applicant has amended claim 1 to even further remove claim 1 from the prior art references

The Neher et al. reference teaches a low glycerol concentration of an aqueous glycerol solution. In other words, the Neher et al. reference teaches a high water content of an aqueous glycerol solution. Claim 1 claims generally the opposite.

As to the Neher et al. reference, the Examiner contends on page 6, lines 7-12 of the Final Office Action that "The Examiner does not consider Applicant's 50 wt % of glycerol substantially different than the art's 40 wt % glycerol. With respect to Applicant's table on page 8 of the Remarks, the Applicant has not compared side-by-side results. The glycerol concentration the Applicant has used is substantially higher than 50 wt %. Thus, the Applicant has not shown a comparison with 50 wt % glycerol, as described in the claim language, with 40 wt % glycerol as taught in the art."

In order to address this contention of the Examiner, applicant has amended claim 1 to revise the water content "not more than 50 % by weight" of the aqueous glycerol solution into --not more than 20 % by weight--. In Example 1 of the Neher et al. reference, a "20 wt. % aqueous glycerol solution [water content: 80 wt. %] is pumped into a vaporizer." While the Neher et al. reference does disclose an aqueous glycerol solution of 10 to 40 wt. % (water content: 60 to 90 wt. %), the Example 1 disclosure appears to be the disclosure in the Neher et al. reference

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having the lowest content of vaporized glycerol which applicant positively recites in claim 1.

It should be noted that the above-amended range --not more than 20 % by weight-- of the water content of the aqueous glycerol solution is also supported by all working examples (Examples 1 to 5) as described in the present application specification (15 % weight, 9 % weight, 8 % weight, 2 % weight, 9 % weight, respectively).

It should be noted that claim 1 claims percentages relative to a water content, while the Neher et al. reference relates its percentages to an aqueous glycerol solution.

In other words, claim 1 claims a water content of not more than 20 % by weight (i.e., 20 % and less than 20 % of water). On the other hand, the Neher et al. reference teaches an aqueous glycerol solution in a range of 10 to 40 wt. % (water content: 60 to 90 wt. %). Hence, there is no overlap between the ranges of claim 1 and the Neher et al. reference.

C.4. If necessary, applicant can submit a declaration

From among all working examples (Examples 1 to 5) as described in the present application specification, there was selected Example 1 where the water content of the aqueous glycerol solution was the highest value 15 % by weight and where the yield of acrylic acid was the lowest value 55 %. Then, in this comparative experiment, acrylic acid was produced in the same way as of Example 1 except that the water content "15 % by weight" of the aqueous glycerol solution was increased to 60 % by weight which was the lower limit value of the water content in the Neher reference.

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As a result, the yield of acrylic acid was 49 %, which was a lower value, even when compared with the yield "55 %" of acrylic acid of Example 1 which yield was the lowest of those of all working examples (Examples 1 to 5) as described in the present application specification.

The result of this comparative experiment demonstrates the superiority of the present invention due to the above-amended range --not more than 20 %" by weight-- over the alleged combination of the Tenten et al. reference with the Neher reference.

D. Housekeeping matters

D.1. Period For Reply

A shortened statutory period for reply was set to expire three months from the mailing date of the Office Action of September 13, 2007. September 13, 2007 plus three months is December 13, 2007. This Amendment and Remarks is being filed on or before Thursday, December 13, 2007 and may be being filed on or before Tuesday, November 13, 2007.

D.2. Status

The Office Action of September 13, 2007 was made final.

D.3. Disposition Of Claims

Claims 1 and 3-6 are pending.

D.4. Application Papers

This case includes no drawings.

D.5. Priority under 35 U.S.C. §§ 119 and 120

Acknowledgement of the claim for foreign priority was made in the Office Action dated May 24, 2007. This is

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appreciated.

D.6. Attachments

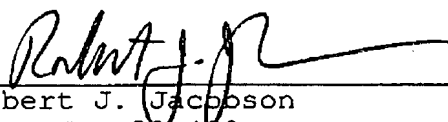
As the Examiner has indicated, applicant has filed one PTO-1449 form in this case. This form has been initialed, signed and returned. This is very much appreciated.

E. Summary

Applicant respectfully submits that the present application is now in condition for allowance. The Examiner is respectfully invited to make contact with the undersigned by telephone if such would advance prosecution of this case.

Respectfully submitted,

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 審査請求 未請求

(全 4 頁)

⑩ アルコールからのオレフィンの製法

⑪ 特 願 昭59—89568

⑫ 出 願 昭59(1984)5月7日

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要 旨

発明の名称

アルコールからのオレフィンの製法

特許請求の範囲

1. 液体又は気体の炭素のアルコールを、このアルコール及び生成するオレフィンの沸点以上の温度で蒸発点範囲中に導入することにより脱水を行い、オレフィンを気体状で抽出し、高沸点炭化を副生物が増加したとき更新し、そして副生物の抽出した高沸点炭化を抽出することを特徴とする、液相中でアルコールから水を分解放出することによるオレフィンの製法。
2. 副生物の増加した炭化を蒸餾することを特徴とする、特許請求の範囲第1項に記載の方法。
3. 高沸点炭化として、ガス油、真鍮ガス油、重質燃料油、工業用白油又は真鍮炭油を使用することを特徴とする、特許請求の範囲第1項に記載の方法。
4. 脱水を、脱水反応に熟知の触媒を用いて行う

ことを特徴とする、特許請求の範囲第1項に記載の方法。

5. 脱水を連続的に行うことを特徴とする、特許請求の範囲第1項に記載の方法。

6. 脱水を150〜550℃で行うことを特徴とする、特許請求の範囲第1項に記載の方法。

発明の詳細な説明

本発明は、高沸点炭化中の炭化で脱水を行い、その炭化を更新して副生物の増加した炭化を抽出に供することによる、アルコールからオレフィンを製造する方法に関する。

炭化の存在下の液相中では、多くの場合アルコールから水がきわめて円滑に分解放出される。それには十分な反応速度に到達するため、後述、例えば脱水又は抽出の高濃度が必要である。脱水反応は通常は液相中150〜200℃で行う。この厳しい条件の結果として、かなり多量の副生物が、例えばエーテル化、異性化及び生成オレフィンの重合により生成する。

オレフィンを得るためのより良好な手段として、兎指中で不斉系触媒を用いてアルロールの脱水が行われる（ハーベン・ウィル著メトペン・アル・オルガニクシエン・ヘミー・Ⅱ 巻 209 頁参照）。触媒として例えば酸化アルミニウム、誘導アルミニウム又はトリウム、チンクマシ及びビチタンの酸化物あるいは複合酸化物が用いられる。反応温度は、 200°C 以上で高いので、特に炭化反応及び炭素の生成が起こりうる。触媒を空気により再生せねばならない。固体触媒を使用する場合の他の欠点は費用の点であり、それは製造においても回収及び抽出に際しても、ならびに廃棄によつても生ずる。兎指反応において合成中不都合の強い物質運搬と供給が困難をきたすので、実用的な熱交換機構及び過熱を考慮せねばならない。

したがつてこれらの欠点を有せず、四生物の簡単な分離検査を可能にする方法を開発することが問題となつていた。

本発明者らは、液体又は気体の状態のアルミ

の同族原子を有するもの。例えばエチノール、
ブチノール又はデカノールがあげられる。

水を介放出する能力のある脂環族のアルコ-
ールは、1個及び多個の化合物例えばシクロヘ-
キサノール、シクロドデカノール、1,2,3,4-テ-
トラヒドロナフトール¹又は1,2,3,4-アト-
ロドデカノール²である。

芳香脂肪族アルコールとしては、例えば2-
フエニルエタノール又は4-フエニルブタノ
ール-1が用いられる。

温水は反応の速順に応じて、触媒なしで又は好ましくは触媒を用いて実施できる。触媒使用の場合、試油に不溶な触媒も触油に可溶な触媒も使用でき、それは触油に溶解され、乳化され又は懸濁される。

好ましくは建築協。例えば屈折族又は劣等族のヌルホシ族例にベンゾールヌルホシ族又はトルオールヌルホシ族、ドプスアベンゾールヌルホシ族、硫酸及び硫酸エニアル例にアルナル族。硫酸又はその錯体はアルナル化された

संख्या 59-210031 (2)

ールを、このアルコール及び生成するオレフィンの沸点以上の温度で高沸點炭化中に導入することにより脱水を行い、オレフィンを生体から排出し、高沸點炭化を微生物が増加したとき更新し、そして微生物の増加した高沸點炭化を排出し、好ましくはこれを無菌に保ち、夜相中でアルコールから脱水することによるオレフィンの製造により、この問題を解決した。

高沸点炭化油とは、沸点150℃を有する高沸点の石油揮発生成物、例えばガスマン、真鍮ガム油、焦油油、工業用油、厚膜ペタフィンワックス又は芳香族炭化水素油を意味する。好ましくは200℃以上の沸点を有する50〜500℃の揮発範囲を有する真鍮ガム油が用いられる。

脱水されるアルコールとしては、反応条件下に安定なオレフィンを生成する、水を分解放出する能力のある低分子のアルコール、特に低分子の飽和性基を有しない、脂肪族、脂環族又は芳香族脂肪族のアルコールが用いられる。

庭園式アルロールの例としては、2〜20個

例導体又は絶縁及びその酸誘導体が用いられる。
 例無水物例えば五硫化磷、二硫化炭素及び硫化
 銅等も使用できる。

炭素不可燃な状態は一般に、炭素に対し0.01~2重量%程度多量しくは0.1~10重量%程度0.5~5重量%の量で炭素に添加される。

熱安定化触媒としては、酸化アルミニウム、硝酸アルミニウム、過化亜硫酸、硫酸アルミニウム、シリカゲル、酸化チタン、炭のヘチマボリ性、ポリブテン酸及びペンタステン酸が用いられる。

風水反用は一般に、50～600で好ましくは50～550で毎分180～250での速度で行われる。一般に大気圧又は加圧が適用されるが、減圧で操作することも可能である。

風水反応用の反応槽としては、例えば攪拌式
 容器が適する。しかし本方法のため特に好まし
 くは、管状に配装された円筒形反応器、例えば
 泡盛等、吹込等又は充填塔等が用いられる。出流
 物質又はその配合物は、通常ガス状で盛油を
 充填した反応器の底部に供給される。あるいは極

状で反応生成物に供給され、その場合気体の供給では反応が、液状で供給される出発物質の沸点よりも高い温度に保持される。気化した出発物質を不活性ガスにより希釈することが有利な場合もある。適当な不活性ガスは、例えば水蒸気、二酸化炭素及び好ましくは酸素である。

反応生成物はガス状で反応器出口から取り出される。次いでガス状の反応生成物を冷却しては凝縮させる。凝縮にさらに精留工程、例えば蒸留又は分留を連続させてもよい。

本発明方法は非連続でも連続でも実行できるが、連続式操作がより優れている。連続式操作では、例えば分解生成物を生ずる反応のときは、気相を連続的に供給しかつ排出することが有利な場合がある。これら分解生成物はその場合液体と一緒に連続的に反応器から排出される。排出された液体の精留処理及び再供給は通常の範囲ではない。なぜならば液体は、例えば燃料油又は気体ガス油として通常設備に容易に入ることができるからである。したがって排出された

例53-210031(S)

分解生成物を含有する原油は連続的に供給し、反応器には新たな原油を供給することが好ましい。

この新規方法は、従来技術の方法に比して次の不利益の利益を有する。

反応器の壁で間に合うか又は熱的な分解も可能であり、そして反応温度は一般に気相中の不均一性による脱水に対するよりも50〜100℃低い。炭素体、分解生成物及び高沸点副産物は原油中に残留し、この原油は再生する必要なしに、場合により反応の分離後、好ましくは発電所に供給される。「油床」中での脱水に因しては、共酸化反応が全く起こらないか又は認めうる程度に起こることがなく、収率が高い。さらに工業的実施における本方法は、文献上しばしば報告されかつ実施されている不均一性による気相反応よりも本質的により経済的である。最後に反応生成物の処理及び原油中に含まれる副産物及び分解生成物は、環境汚染を減らすことができる。

実施例1

装置は、ガラス瓶(容量264、気体ガス相(沸点400℃)14g及び5gのドデシルベンゾールとメチルベンゼンを含有した反応器(容量600ml、長さ1100mm)から成る。反応器を200℃で予熱した後、同時に空気を25gと導入する。反応器から出る反応混合物を40℃で凝縮させると、液体725gが得られる。このものは反応プロダクト97重量%、水22重量%及びイソブタン42重量%から成る。これは90.5%の収率に相当する。イソブタンの収率は、反応した反応プロダクトに対して理論値の97%である。

実施例2

実施例1の装置と同様に操作し、ただし反応温度を165℃に保つ。三級ブタノール14g、水20.0重量%及びイソブタン62.9重量%を含有する原油が72.7g得られる。

これは84%の収率に相当し、収率は理論値の97%である。

実施例3

実施例1に記述の装置に、215℃で毎時25gと一様にシクロヘキサノール100gを供給する。同時に原油724gが得られ、このものはシクロヘキサノール24重量%、シクロヘキサノール15重量%、水13.8重量%、シクロヘキサノール74.4重量%から成る。これは95%の収率及び理論値の96%の収率に相当する。

実施例4

実施例1に記述の装置に、250℃で毎時ドデシルベンゾールとメチルベンゼン60重量%と一様に反応ガス相1200重量%中のブタノール1の51重量%を導入する。毎時ブタノール1の44.5重量%、水13.8重量%、ブタノール1の36.2重量%、トランス-2-ブタン2.8重量%及びシス-2-ブタン2.4重量%からの原油50重量%が得られる。これは反応した

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ブタノール-1 に対し 5 6 % の変化率及びブタ
ノール-2 8 6 %、トランス-2-ブタノール 7 % 及び
シス-2-ブタノール 6 % の収率に相当する。

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English translation of selected passages from JP-A-59-210031 (Kokai)

Page 1, lower left column, lines 4-12 (claim 1):

Claims:

- 5 1. A process for producing an olefin by decomposition-extrusion of water from an alcohol in a liquid phase, which is characterized by comprising the steps of: carrying out a dehydration reaction of an alcohol by introducing the alcohol in a liquid or gaseous state into a high-boiling-point mineral oil at a temperature not lower than a boiling point of the alcohol and a boiling point of an olefin being formed from the
- 10 alcohol; discharging the formed olefin in a gaseous state; renewing the high-boiling-point mineral oil when by-products have increased; and discharging the high-boiling-point mineral oil having the increased by-products.

Page 3, lower left column, line 1 to lower right column, line 2 (Examples 1 and 2):

15 Example 1

An apparatus comprises an electric heating type quartz tube (D = 60 mm, L = 1100 mm) filled with 2.6 liters of glass beads (D = 5 mm), 1.4 kg of a vacuum gas oil (boiling point 400 °C) and 5 weight % of dodecylbenzenesulfonic acid. An amount of 74.1 g/h of tert-butanol is supplied through an immersion tube at 205 °C by a

20 metering pump. At the same time, 2.5 liters/h of nitrogen is introduced. If a reaction mixture discharged from the reactor is condensed at -40 °C, then 72.55 g of a condensate is obtained. This condensate comprises 9.7 weight % of tert-butanol, 22.5 weight % of water and 67.8 weight % of isobutene. This corresponds to a conversion of 90.5 %. The yield of isobutene is 97 % of a theoretical value based

25 on the reacted tert-butanol.

Example 2

The same operation as Example 1 is carried out except that the reaction temperature is kept at 165 °C. An amount of 72.7 g of a condensate comprising

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16.3 weight % of tert-butanol, 20.8 weight % of water and 62.9 weight % of isobutene is obtained. This corresponds to a conversion of 84 %. The yield is 97 % of the theoretical value.